PB82-126319

Tethered Balloon Sampling Systems for Monitoring Air Pollution

Denver Research Inst., CO

Prepared for

Industrial Environmental Research Lab.
Research Triangle Park, NC

1981

U.S. Department of Commerce
National Technical Information Service
NTIS.

EPA-600/J-81-383
Journal Article

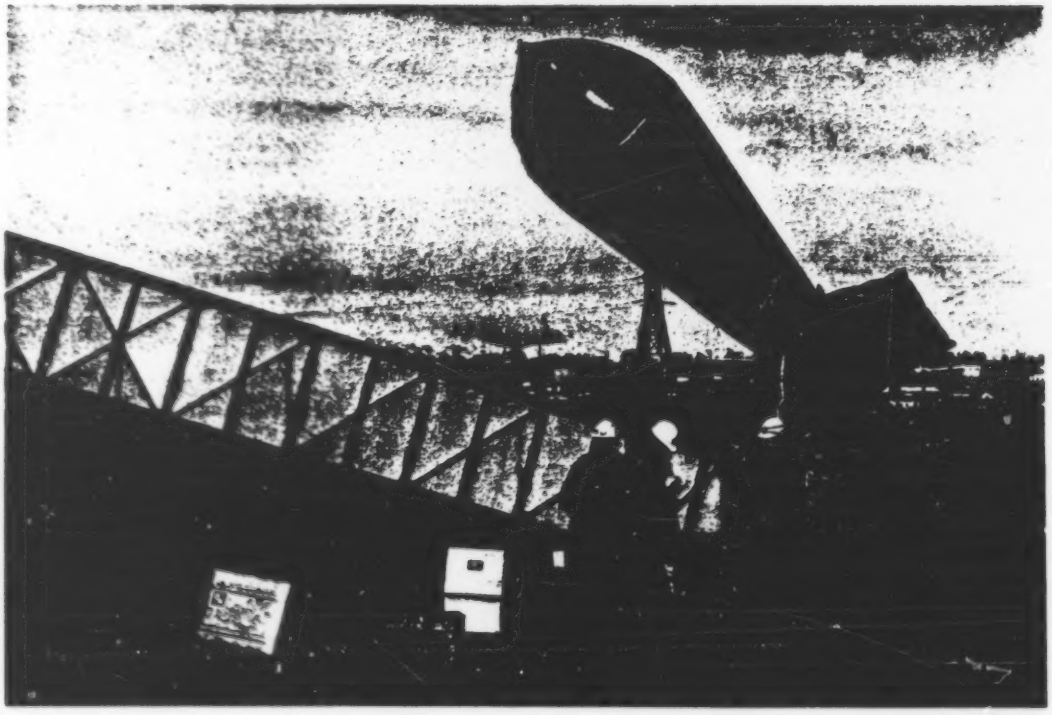
July 1981
Volume 31, Number 7
JPCAAC 31(7) 717–822 (1981)

PB82-126319

JOURNAL OF THE AIR POLLUTION CONTROL ASSOCIATION

JAPCA

Tethered balloon sampler being prepared for plume flyash collection



735 Tethered balloon sampling systems

744 FGD waste treatment and disposal

808 Air quality control equipment guarantees

REPRODUCED BY
NATIONAL TECHNICAL
INFORMATION SERVICE
U.S. DEPARTMENT OF COMMERCE
SPRINGFIELD, VA 22161

TECHNICAL REPORT DATA
(Please read Instructions on the reverse before completing)

1. EPA-600/J-81-383	2. Journal Article	3. RECIPIENT'S ACCESSION NO. PB82 126319
4. TITLE AND SUBTITLE: Tethered Balloon Sampling Systems for Monitoring Air Pollution		5. October 1981 6. PERFORMING ORGANIZATION CODE 310032
7. AUTHOR(S): James A. Armstrong/Philip A. Russell (DRI, U. of Denver) and Leslie E. Sparks/Dennis C. Drehmel (EPA/IERL-RTP)		8. PERFORMING ORGANIZATION REPORT NO.
9. PERFORMING ORGANIZATION NAME AND ADDRESS: Denver Research Institute, University of Denver, Denver, Colorado		10. PROGRAM ELEMENT NO. 11. CONTRACT/GRANT NO.
12. SPONSORING AGENCY NAME AND ADDRESS: **EPA, Office of Research and Development, Industrial Environmental Research Laboratory, Research Triangle Park, NC 27711**		13. TYPE OF REPORT AND PERIOD COVERED: Journal Article; 1981 14. SPONSORING AGENCY CODE: **EPA/600/13**

15. SUPPLEMENTARY NOTES: Published in the Journal of the Air Pollution Control Association, Vol. 31, No. 7, pp. 735-743, 1981.

16. ABSTRACT: The paper is an overview of recent studies in which balloons, usually tethered, have been used to investigate the structure and air quality of the planetary boundary layer. It also describes a number of lightweight tethered balloon sampling systems, developed to investigate particulate and gaseous emissions from point and nonpoint sources. The sampling systems have both vertical and horizontal mobility and sample at specified altitudes for extended periods of time. Two of the balloon-borne samplers are remote controlled and have been used to monitor emissions in the plumes of point sources. A third sampler has been developed to investigate the vertical extent and downwind transport of particle emissions from fugitive sources. A fourth sampler, currently being developed, is also described.

17. KEY WORDS AND DOCUMENT ANALYSIS

a. DESCRIPTORS	b. IDENTIFIERS/OPEN ENDED TERMS	c. COSATI Field/Group
Pollution	Pollution Control	13B 13H
Processing	Stationary Sources	01C 14G
Tethered Balloons	Particulate	14B
Leakage	Fugitive Emissions	11G
Sampling		07D
Dust		
Aerosols		21B
Gases		
Plumes		

18. DISTRIBUTION STATEMENT: **Release to Public**	19. SECURITY CLASS *(This Report)*: **Unclassified**	21. NO. OF PAGES: 9
	20. SECURITY CLASS *(This page)*: **Unclassified**	22. PRICE

EPA Form 2220-1 (9-73)

TETHERED BALLOON SAMPLING SYSTEMS FOR MONITORING AIR POLLUTION

James A. Armstrong and **Philip A. Russell**
Denver Research Institute
University of Denver
Denver, Colorado

Leslie E. Sparks and **Dennis C. Drehmel**
U. S. Environmental Protection Agency
Industrial Environmental Research Laboratory
Research Triangle Park, North Carolina

An overview of recent studies in which balloons, usually tethered, have been used to investigate the structure and air quality of the planetary boundary layer is presented. Also described are a number of lightweight tethered balloon sampling systems which have been developed to investigate particulate and gaseous emissions from point and nonpoint sources. The sampling systems have both vertical and horizontal mobility and are capable of sampling at specified altitudes for extended periods of time. Two of the balloon-borne samplers are remote controlled and have been used to monitor emissions in the plumes of point sources. A third sampler type has been developed to investigate the vertical extent and downwind transport of particle emissions from fugitive sources. A fourth sampler, currently being developed, is also described.

Air pollution and ambient air quality studies are usually confined to the earth's planetary boundary layer, which is a zone extending from the surface to the free atmosphere where surface frictional effects no longer apply. This boundary layer typically reaches to an altitude of about one kilometer. Planetary boundary layer investigations can be separated into two broad categories: atmospheric structure studies and air quality studies. The latter category can be further subdivided into dispersion studies and atmospheric constituent studies. The atmospheric structure studies are concerned with measuring properties of the boundary layer, such as the turbulent transport of heat and momentum. Dispersion studies are concerned with how natural and man-made pollutants are transported and diffused in the environment. Atmospheric constituent studies involve measuring the concentrations and compositions of the pollutants.

Since the first recorded manned balloon-borne observations took place on September 30, 1784, when a hot air balloon was used to make vertical measurements of atmospheric pressure, temperature, and humidity,[1] balloon systems have been employed to study the planetary boundary layer. This paper provides an overview of recent studies in which balloons, usually tethered, have been used to investigate the lower strata of the atmosphere and also describes new lightweight tethered balloon sampling systems developed to monitor particulate and gaseous emissions from point and nonpoint sources and urban air masses.

Overview of Past Balloon Sampling Investigations

The planetary boundary layer structure has been investigated by several researchers using instruments carried aloft by tethered balloons. Readings and Butler[2] have described the development and testing of turbulence probes that attach to the tetherline of a captive balloon. The probes measure wind and temperature fluctuations. Haugen, *et al.*[3] used these probes in conjunction with a blimp-shaped 1275 m^3 tethered balloon to determine momentum and heat fluxes in the planetary boundary layer. Vertical profile measurements were made to heights of 1200 m using five probes spaced on the tetherline. The field program showed that turbulent transport measurements can be made directly using an eddy correlation technique.

A much smaller tethered balloon sounding system has been described by Morris, *et al.*[4] This system used a blimp-shaped balloon having a buoyant gas capacity of 3.25 m^3. The system's radio transmitting package measured pressure, temperature, wet-bulb temperature, wind speed, and wind direction. The system was highly portable; could be operated in winds of 10 m/s; and could reach altitudes of 750 m in winds up to 6 m/s. This system has been used by Whiteman and McKee[5,6] to study the vertical temperature and wind structures in a deep mountain valley. Observations of inversion break-up within the valley were made and a hypothesis to account for the observations has been developed. The tethered balloon was operated to a height of 450 m.

An investigation of the dispersion of fluorescent particle tracers released upwind of an isolated urban area has been reported by Hilst and Bowne.[7] The enhancement of vertical mixing over the city due to increased surface roughness and thermal effects was established as compared to nearby rural areas. In addition to an extensive ground network of rotating impaction type samplers used to measure surface dosages of the released tracer material, the field program employed additional samplers on a 450 m balloon tether and on a 244 m broadcasting tower to monitor the vertical dosages. (As in several papers that follow, specific information on the balloon used in this study was not presented.) Vertical wind and temperature measurements were made during the tracer studies using wiresondes and a modified radiosonde.

A series of four rotating impaction samplers connected to the tetherline of a 14 m^3 balloon was used by Mandrioli and Tampieri[8] to investigate atmospheric transport processes. They measured the vertical profiles of pollen and spore concentrations to maximum height of 800 m during convective conditions and compared the observations with turbulent transport models. Temperature and pressure soundings were made during sampling using a radiosonde.

A number of researchers have used tethered balloon systems to sample gaseous constituents of the atmosphere. Galbally[9] employed a Mast-Brewer ozonesonde suspended from a tethered balloon to study the variations of O_3 concentrations of a remote area. The concentrations were measured at selected heights up to 200 m over 24 hr periods. The observed O_3 destruction rates were compared to other measurements. Ozone concentrations to 1500 m were measured by Sekihara, *et al.*,[10] using an ozonesonde attached to a tethered balloon. They also employed ozonesondes at the surface and at the 200 m level on a tower. They report a number of vertical profiles and time cross-sections of O_3 concentration and temperature for different atmospheric conditions.

A radiosonde and three air samplers, carried by a single tethered balloon, has been developed by Cleeves, *et al.*[11] for monitoring urban plumes. The blimp-shaped balloon had a free lift of 8.5 kg and was operated to heights of 300 m. The radiosonde measured temperature and humidity; the air samplers collected grab samples for later laboratory analysis using gas chromatography. A field program conducted over a city is described in which temperature and humidity soundings are presented as well as mean methane concentrations collected simultaneously at heights of 30, 60, and 275 m during stable atmospheric conditions.

Air sampling in the vicinity of a chemical plant has been reported by Rohrschneider, *et al.*[12] Three sampling devices were spaced along the tetherline of a 65 m^3 spherical balloon, which had a lifting capacity of 15 kg. The sampling devices utilized liquid nitrogen to condense the air samples. The air samples were analyzed by gas chromatography for C_3 to C_6 hydrocarbons, methyl chloride, and vinyl chloride. The results indicated that the concentrations of the above gases decreased to half of the surface concentrations at a height of 200 m.

Blimp-shaped captive balloons, varying in size from 40 to 1500 m^3, have been used by Perroud, *et al.*[13,14] to investigate gaseous pollution in the lower layers of the atmosphere. The authors presented the operating characteristics of the various balloons and discussed the sampling devices which attached to the tetherlines. A radio probe was used to obtain soundings of pressure, temperature, humidity, and wind velocity. Air sampling devices included liquid nitrogen traps for hydrocarbon determination (gas chromatography) as well as bubblers for SO_2 sampling (West-Gaeke method) and chlorine sampling (colorimetric method). Results were reported showing the effect of temperature inversions on pollution levels.

Garland and Branson[15] used five samplers attached to the cable of a tethered balloon to measure the SO_2 concentration near an industrial area. The measurements were made at heights from 150 to 1200 m. The samplers drew air through bubblers containing a solution of hydrogen peroxide which reacted with the SO_2 to form sulfate. The measurements indicated that the SO_2 mean mixing height was ~1200 m. The paper also discussed the SO_2 mass balance of the area's atmosphere.

A tethered balloon system for profiling smoke plumes from forest fires and other open combustion sources to an altitude of 600 m has been developed by Ryan, *et al.*[16] Twelve sampling packages, which monitor temperature and wind speed and sample particulate matter and combustion gases, were attached to the tetherline of a 99 m^3 blimp-shaped balloon having a lift capacity of 106 kg. Particles were collected by drawing air at a known flow rate through Teflon or glass fiber filters. The filters were analyzed gravimetrically. Combustion gases were collected in grab bags. CO_2, CO, and NO_x concentrations were analyzed on site using nondispersive infrared, electrochemical, and chemiluminescence techniques. The paper described the field operations and a series of demonstration experiments.

Vertical measurements of temperature, humidity, wind speed, and aerosol concentrations to heights of 500 m have been made by Sentell, *et al.*[17] The measurements were made during an aircraft noise propagation study. A 42 m^3 blimp-shaped balloon was employed. The aerosol counter was a photoelectric forward scattering instrument capable of detecting particles ≥0.3, ≥0.5, and ≥3.0 μm in diameter. The vertical soundings obtained over a 4-hr period were reported. Using the same aerosol counter system, Rosen and Hofmann[18] have investigated the near surface aerosol properties in an arid region. They employed a 70.8 m^3 tethered blimp-shaped balloon and made measurements to 1.7 km. A free balloon was also used to obtain meteorological soundings. The investigation was conducted to obtain data for optical atmospheric transmission models. The measurements showed that, during the springtime at the test site, the surface concentration of aerosols between 0.3 and 3.0 μm in diameter was representative of the concentrations monitored to heights of ~1 km.

Two manned free balloon systems have been recently developed for air quality monitoring. A superpressure balloon has been described by Heinsheimer[19] which is capable of tracking a parcel of air for over 30 hr. The spherical balloon, called an ATMOSAT for atmospheric satellite, is 10 m in diameter and has been operated to altitudes of 2500 m. The paper described the first four flights of the balloon. In future flights, the author suggests that the balloon be used to measure O_3, NO_x, SO_2, temperature gradients, turbulence, etc.

Measurements of the conversion of SO_2 to SO_4^{-2} over an urban area has been made by Forrest, *et al.*[20] using an instrumented manned balloon called the Da Vinci. The 20 m diameter balloon carried a crew of four and one metric ton of instrumentation in a suspended gondola. During two flights a tandem filter sampling system was used to collect simultaneously particulate sulfate and SO_2 for periods of 2 hr as the balloon tracked urban air masses. The results indicated extremely slow oxidation rates ranging from 0 to 4%/hr.

To conclude this overview of planetary boundary layer investigations utilizing balloon systems, two reports summarizing surveys of the user community are listed. In 1975, Baynton, *et al.*[21] conducted a survey on scientific levels of interest in blimps and tethered balloons for the NCAR Research Aviation Facility Advisory Panel. Kuhner and McDowell[22] reported the results of a 1978 survey sponsored by NASA on user interests involving five "strawman" airborne platforms. Two of the strawman platforms, a powered mini-blimp and a large tethered balloon, are suitable for a wide variety of boundary layer investigations.

Development of Lightweight Balloon Sampling Systems

With the exception of the balloon sounding system described by Morris, *et al.*,[4] the tethered balloons used in past planetary boundary layer investigations would be subject, due to their sizes, to FAA regulations[23] if operated in the United States. Unless waivers could be secured, the operating limitations of the regulations would severely restrict the use of these balloons. The horizontal mobility of large balloon systems is also limited. Besides federal regulations, the actual size of a tethered balloon restricts where it can be safely and conveniently operated, e.g. see Ryan, *et al.*[16] Finally, the capital and operating costs of large balloon systems can also be prohibitive. In general, large balloons require larger field operating crews which at times will be idled by inclement weather.

With the above limitations in mind, the Denver Research Institute (DRI), with EPA support, has developed various types of lightweight samplers which are carried aloft by tethered balloons of a size not restricted by FAA regulations. The tethered balloon sampling systems: (1) possess both horizontal and vertical mobility; (2) are capable of collecting long term samples; (3) can safely collect samples in close proximity to emission sources; (4) can simultaneously collect particulate and gaseous samples; (5) are relatively easy to operate; (6) provide samples suitable for multidisciplinary analysis; and (7) are inexpensive.

To date, three types of lightweight battery operated balloon samplers have been designed, fabricated, and utilized. A fourth sampler type is currently under development. The first sampler to be developed is remote controlled and is capable of collecting either a number of discrete particle samples or a time resolved "streak" sample on a linear filter substrate. Treated filters can be used with this sampler for monitoring some gases. An air sampling bag can also be employed. The second sampler is also remote controlled and is designed to collect a number of discrete particle samples on individual filter substrates. In addition, this sampler can be used to take a number of gas samples by means of adsorption tubes, colorimetric detection tubes, midget impingers, air sampling bags, etc. Both remote controlled samplers were originally designed and have been employed to conduct plume sampling of fly ash from coal-fired power plants where conditioning agents were being evaluated.[24,25] These samplers have also been used to collect aerosols from a Portland cement plant which was experiencing problems with a persistent detached plume.[26] The simultaneous collection of particulate and gaseous samples has been successfully tested using the remote controlled samplers. Gas sampling has yet to be employed, however, during an operational field program.

The third sampler type to be developed is extremely lightweight and wind directional. The sampler is not remote controlled and collects a single sample. Three of these samplers, however, can be flown simultaneously to collect airborne particles at different selected elevations using one balloon system. These samplers have been designed to investigate the vertical extent of fugitive dust emissions from various operations at a surface coal mine.[27]

A fourth sampler, which is also extremely lightweight and wind directional, is currently being developed to collect the inhalable particulate (IP) fraction of airborne particles from fugitive sources. Inhalable particles are currently defined by EPA as those having an aerodynamic diameter of 15 μm or less.[28] The new sampler will utilize horizontal elutriation to obtain a 15 μm aerodynamic diameter cut and will be capable of sequential filtration to provide a second cut to separate the fine and coarse size fractions of the inhalable particles sampled. It is planned to use a number of these samplers to characterize IP emissions from line, point, and area fugitive sources.[29] The sampling method will consist of using a series of tethered balloon systems positioned at selected distances downwind from the source being sampled. Three samplers will be flown simultaneously at different heights by each balloon system with a fourth sampler located at ground level at each launch site. The multiple balloon sampling technique will be coupled with the exposure profiling method developed by Midwest Research Institute[30] to determine improved source emission factors. The balloon network will also be used to characterize the downwind transport of fugitive emissions.

The major components of the tethered balloon sampling systems are discussed in the sections that follow.

Tethered Balloon System

A commercially available 3.25 m^3 blimp-shaped balloon is used as the airborne platform for the samplers. When inflated with helium, this balloon has a static lift of 1.9 kg at sea level. The balloon is polyethylene and, for safety and ease in observation is red. The balloon is attached by a tetherline to a battery-powered portable winch that weighs 27 kg. The winch contains a forward/reverse, variable speed motor that drives the tetherline spool and a 12 V sealed lead/acid battery pack and charger system. The extremely lightweight tetherline has a breaking strength of 535 N and a mass to length ratio of 0.4 kg/km.

This balloon system is capable of lifting a package weighing 1.2 kg to maximum altitudes of 800 to 1000 m at sea level in winds of 6 to 8 m/s. Flight performance of the balloon is still good in winds of 10 m/s but the maximum altitude attainable is somewhat reduced. The balloon is designed to survive in winds up to 20 m/s.

Meteorological Package

A commercially available battery operated meteorological package is attached to the balloon or tetherline during sampling. The meteorological package, which weighs 135 g, constantly measures and telemeters pressure, temperature, and wet bulb temperature to a recording ground station. The relative humidity of the air being sampled and the height of the balloon above the ground during flight operations can be calculated using these data.

First Generation Remote Controlled Sampler

This sampler consists of a movable sampling head connected by flexible tubing to a piston pump and a flow-adjust needle-valve loop followed by a flow meter. A nozzle attached to the sampling head translates along a filter strip which is used to collect airborne particles when air is drawn through it. The sampling head is translated by means of a guide mechanism consisting of a guide rod and a motor driven lead

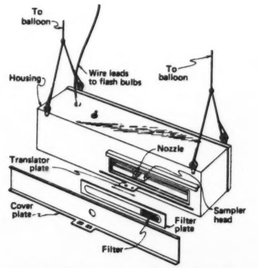


Figure 1. First generation remote controlled sampler.

Air Pollution Control Association

screw. The sampler contains a radio receiver/servo system used to control selectively, from the ground, the sampling pump, the leadscrew motor, and a flight termination system which deflates the balloon in case of a tetherline failure. The sampler also contains a radio transmitter system used to verify that the sampler is operating correctly. The total weight of this package is 1050 g.

A schematic showing the front perspective view of the sampler is presented in Figure 1. The overall dimensions of the sampling package are 43 × 11.4 × 8.9 cm. The sampler housing consists of a rectangular box of thin aluminum sheeting. Aluminum, plastic, and nylon are used extensively throughout the package to minimize weight. The means of suspending the sampler to the balloon is also shown in Figure 1. The two lines that attach to the balloon are on opposite sides of the balloon body. Since the balloon points into the wind, this method of attachment gives the operator the flexibility of orienting the sampler so that the filter is either pointing into or away from the wind.

The filter strip is mounted to a slotted aluminum plate which fits into a rectangular aperture in the front wall of the sampler housing. A filter cover plate protects all but the active sampling portion of the filter. A 0.4 μm Nuclepore substrate is normally attached to the metal filter holder by an appropriate adhesive. The nozzle that comes in contact with the Nuclepore filter has a sampling area of 0.7 cm^2 through which the pump is capable of drawing air at a maximum rate of 1.5 L/min. Other filter media, such as Millipore or glass fiber, are also compatible with this sampler.

A ground-based radio transmitter is used to send command signals to the sampler receiver. The receiver controls servos for flight termination, filter sample position, and pump actuation. The flight termination servo circuit is wired to a flashbulb assembly attached to the balloon skin. In case of a tetherline failure, the ground operator can activate the bulbs to burn a hole in the balloon skin, causing the balloon to descend slowly to the ground.

The translate servo controls the actuation of the leadscrew motor which changes the location of the sampling head relative to the filter. When the sampling head reaches a new filter location, a limit switch deactivates the leadscrew motor. The balloon system operator is informed by telemetry when the sampling head has translated to a new sampling position.

The pump servo controls the actuation of the sampling pump. Pump operation is also verified by telemetry.

The battery pack consists of four series-wired lithium cells which provide 11.2 V for operating the transmitter. A tap on the battery pack at 8.4 V is regulated to provide a fixed 5V for operating the sampling pump, receiver, servos, and verify circuit. The battery pack powers this sampling package for a maximum of 10 hr. The voltage level of the battery pack is constantly monitored and, when it falls below a set value, a signal is telemetered to the operator indicating that the unit must be replaced.

This sampler can also be operated in a time-resolved "continuous streak" sample mode. This requires that a slit sampling head be used as well as a gear box attached to the leadscrew motor which causes the sampling head to translate across the filter at a selected reduced speed. After positioning the balloon-borne sampler in the desired location the ground operator simply actuates the sampling pump and the translate mechanism.

Finally, a treated filter or an air sampling bag can be used with this package to sample gases. The air sampling bag would be connected to the pump outlet.

Second Generation Remote Controlled Sampler

This sampler is based on a modular concept and weighs 950 g. One configuration of this sampling package, shown in Figure 2, permits the use of filters to collect particles or air sampling bags to sample gases. This configuration consists of a "base module" and a "rotating valve module." The base module contains a radio receiver, two servos, a verify circuit, a verify transmitter, a voltage regulator, a piston pump, a battery pack, and electronics for a gas sensor. These components are encased in polystyrene foam. Thin-walled aluminum rods connect the base module to the rotary valve module. The dimensions of this sampling package are 35 × 15 × 15 cm.

The rotary valve module contains an 8-position nylon rotary valve containing an O-ring seal. The valve rotor is driven by a low speed gear motor. The location of the rotor vs. stator valve ports is controlled by a limit swtich. Flexible tubing connects the rotary valve stator ports to eight 37 mm filter cassettes and the valve rotor port to the sampling pump. The valve/motor assembly and filter holder are attached to a thin aluminum plate.

As with the first generation remote controlled sampler, the radio receiver of this package receives command signals from a ground-based transmitter. The receiver is coupled to servo circuitry consisting of a flight termination servo and a valve motor/pump motor servo. An onboard verify transmitter circuit is again used to alert the ground operator that the commands have been received by the sampler and have been acted upon correctly.

The gas detector is a heated surface semiconductor device which is sensitive to various hydrocarbon and reactive inorganic gases. The output of this detector is interfaced to a log amplifier which, in turn, is interfaced to the balloon-borne transmitter. By this means, continuous relative information concerning the placement of the balloon in the envelope of the plume being sampled is transmitted to the ground-based receiver.

Four lithium cells again make up the battery pack for this package. As with the first generation sampler, the cells are wired in series to provide 11.2 V to operate the transmitter. A voltage regulator again provides a fixed 5 V from an 8.4 V tap for operating the gas detector, sampling pump, receiver, servos, and verify circuit. With 5 V applied to the pump, a constant flow rate of 2.5 L/min is drawn through a 0.4 μm Nuclepore filter whose effective diameter is 33 mm. Millipore and glass fiber filters are also used to collect particle samples. The battery pack is capable of operating the sampling package for a maximum of 7 hr.

Air sampling bags can also be used with this package by reversing the connection of the flexible tubing from the pump inlet to the pump outlet. The sampling bags are attached to the exterior of the sampler by rubber bands and the bag filling lines are connected to the eight stator ports.

A second configuration of this sampling package involves adding a "gas sampling" module to the front of the rotary valve module. This extension module is used for the addition of a matrix of gas sampling components such as adsorption tubes, colorimetric detection tubes, and midget impingers. This design permits simultaneous sampling of gases and particles.

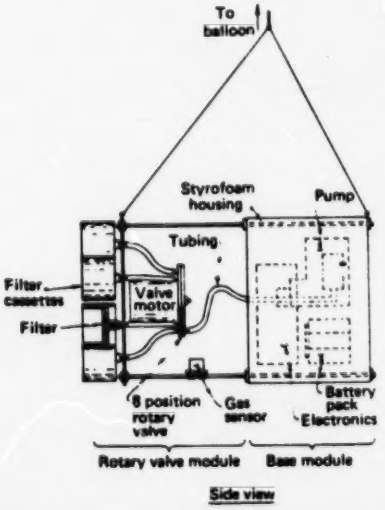


Figure 2. Second generation remote controlled sampler.

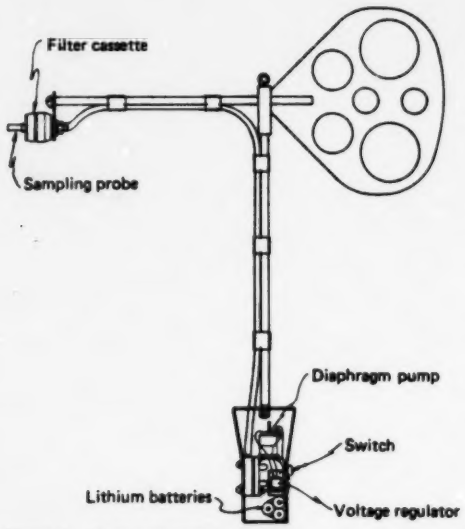


Figure 3. Wind vane particle sampler.

Wind Vane Particle Sampler

A schematic of this sampler is shown in Figure 3. Three of these samplers can be carried simultaneously by the balloon system. Each sampler weighs 400 g. The vertical tube of the sampler attaches to the horizontal tube at a point where the wind vane balances the forward tube extension such that the extension is always horizontal. The tubes and wind vane are fiberglass. A 37 mm filter cassette is attached to the front end of the horizontal tube. An assembly housing the pump, battery pack, and voltage regulator is attached to the bottom of the vertical tube. The weight of these components ensures that the sampler maintains the orientation shown in Figure 3. Nylon lines equipped with swivels and quick disconnect clamps are used to attach the samplers to the balloon and tetherline. The wind vane configuration ensures that the samplers point into the wind during sampling. Observations have shown this to be the case even in very light winds (<1 m/s).

The samplers are equipped with diaphragm pumps which are powered with battery packs consisting of three lithium cells wired in series which provide 8.4 V. This voltage is regulated down to 5 V to ensure constant pumping rates. Flexible tubing connects the pumps to the 37 mm filters held in plastic cassettes. Each sampling pump is activated at the time that the associated sampler is launched. When 0.4 μm Nuclepore filter substrates are employed, the air sampling rate is 4.75 L/min. Millipore filters having an effective pore size of 0.8 μm are also used, resulting in an air flow rate of 5.1 L/min. The sampler can operate for 10 hr before the battery pack needs replacement.

Sampling probes were also developed for the 37 mm filter cassettes. The probes, which are 2.5 cm long and have an ID of 4 mm, were designed so that isokinetic sampling would exist in winds of approximately 6 m/s.

IP Wind Vane Sampler

This sampler, currently under development, will use a lighter weight version of wind vane and tubing arrangement of the previous sampler. The tubes will still be fiberglass; the wind vane will consist of aluminized Mylar sheets stretched over a wire loop. In addition, this device will have a particle sampling assembly consisting of a fiberglass entrance cone, a horizontal metal tube elutriator, and a polystyrene cassette capable of holding one or two filters. Figure 4 shows the particle sampling assembly in detail.

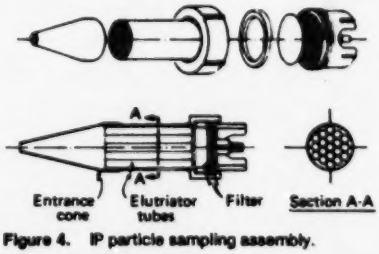


Figure 4. IP particle sampling assembly.

The assembly will attach below the horizontal tube extending from the wind vane (see Figure 3). This sampler will use a diaphram pump, a lithium battery pack and a 5 V regulator assembly similar to the previous sampler. In addition, a needle valve will be used to bleed air into the flexible tube connecting the sampling assembly to the pump. This will allow for fine control of the airflow through the sampling assembly. The sampling assembly is being designed for a constant flow rate of 2 L/min.

The horizontal elutriator will consist of a matrix of 19 thin wall stainless steel tubes, each having a radius of 2.3 mm and a length of 3.5 cm. For the above flow rate, the elutriator has a calculated aerodynamic diameter particle cut point of 15 μm.[29] The 19 tubes will be housed in a thin wall aluminum tube having an outside diameter of 2.5 cm. Interstitial spaces between adjacent elutriator tubes will be filled with epoxy.

Sequential filtration will be used to separate the fine and coarse size fractions of the particles exiting the elutriator. Stacked 25 mm diameter Nuclepore filters will be used where the first filter, having a pore size of 8.0 μm, will collect the coarse particles and the second filter, having a pore size of 0.3 μm, will collect the fine particles. (The filter cassette in Figure 4 shows only the base section of the commercially available filter assembly. This configuration would be used when sequential filtration is not employed.) By controlling the face velocity of air passing through the filters, the Air Quality Group at the University of California, Davis[31] has shown it is possible to obtain a good size cut between the coarse and fine particles. The actual filtration area of the 25 mm filters is 2.4 cm^2 so for an air flow rate of 2 L/min, the face velocity would be 14 cm/s resulting in a cut point of ~2.3 μm. The outer cassette cap will be permanently attached to the back of the elutriator housing tube.

The diverging entrance cone of the particle sampling assembly will have a total included angle of 30° or less to ensure that separation from the walls will not occur as the air is decelerated prior to entering the elutriator. The cone will slip over the front of the elutriator housing tube. To approach isokinetic sampling in winds of 2 to 10 m/s, three interchangeable cones having different inlet diameters will be used.[27] The decision of which cone to employ will be dictated by the average wind speed encountered at the start of a sampling period.

A prototype IP wind vane sampler has been fabricated and is currently undergoing laboratory testing. The prototype sampler weighs 350 g.

Field Operations

A field crew of two people is usually required to launch, operate, and retrieve the tethered balloon sampling systems. A typical field operation at a point source emitter, such as a coal-fired power plant, consists of first inflating the balloon and then transporting the balloon, winch, sampling equipment, and meteorological station to a launch site at a selected distance directly downwind of the stack to be sampled. Next, the balloon is attached to the winch tetherline and the meteorological

package and a remote controlled sampler are attached to the balloon. Prior to balloon launch, the meteorological package and ground station are activated and temperature, wet bulb temperature, and pressure readings are recorded for the ground location. The above prelaunch activities can be accomplished in approximately 30 min.

When sampling operations take place at downwind distances of 300 m or less from a power plant stack, proper vertical positioning of the balloon in the plume is easily accomplished based on visual ground observation. Experience has shown that during ascents into visible plumes, the horizontal motion of the balloon is usually minimal until the balloon enters the plume envelope. Once in the plume, the horizontal tracking motion of the balloon matches the visible smoke motion of the plume passing by the balloon. Furthermore, when a visible plume is sampled, a portion of the plume can always be seen between the balloon and the observer. The gas detector of the second generation remote controlled sampler has also proved to be useful in determining the envelope of less visible plumes. Tests conducted in plumes of both a coal-fired power plant and a portland cement plant have confirmed that the detector is capable of informing the ground operator when the balloon is in the plume of a point source. Once the sampler is properly positioned in the plume, the ground operator activates the sampler via radio control and sampling commences. During sampling, temperature and pressure information from the balloon-borne meteorological package is periodically recorded.

Particle sampling times vary depending on the filter loadings needed for specific analyses, the concentration of the plume being sampled, the downwind sampling distance, and the type of sampler being used. Experience has shown that, when plume sampling is conducted 150 m downwind from a power plant stack, 5 min sampling times are adequate for collecting sufficient fly ash for microscopic analyses while 30 min sampling periods provide sufficient filter loadings for bulk elemental analyses. During one field program in which a power plant plume was sampled 900 to 1000 m from the stack, 20 min sampling times were sufficient for microscopic analyses and 120 min times produced loadings sufficient for bulk analysis. Results of these studies are reported elsewhere.[24,25]

The wind vane particle samplers were first used during a field program to monitor the height of fugitive dust emissions from activities at a surface coal mine. Field operations consisted of locating the balloon system at a selected distance downwind of the mine operation(s) to be sampled. The first sampler was then attached to the balloon and the pump of this sampler was activated. The balloon was launched and a selected length of tetherline was reeled off the winch. The winch was stopped, the second sampler attached to the tetherline, and its pump activated. Again the winch was started and the appropriate length of tetherline reeled out. The winch was stopped, the third sampler attached to the line, and its pump activated. Finally, additional tetherline was run out until the samplers reached the desired sampling heights. (Appropriate distances had been marked on the tetherline to assist in placing the samplers at the desired heights.) A fourth sampler, near the winch, collected particles at ground

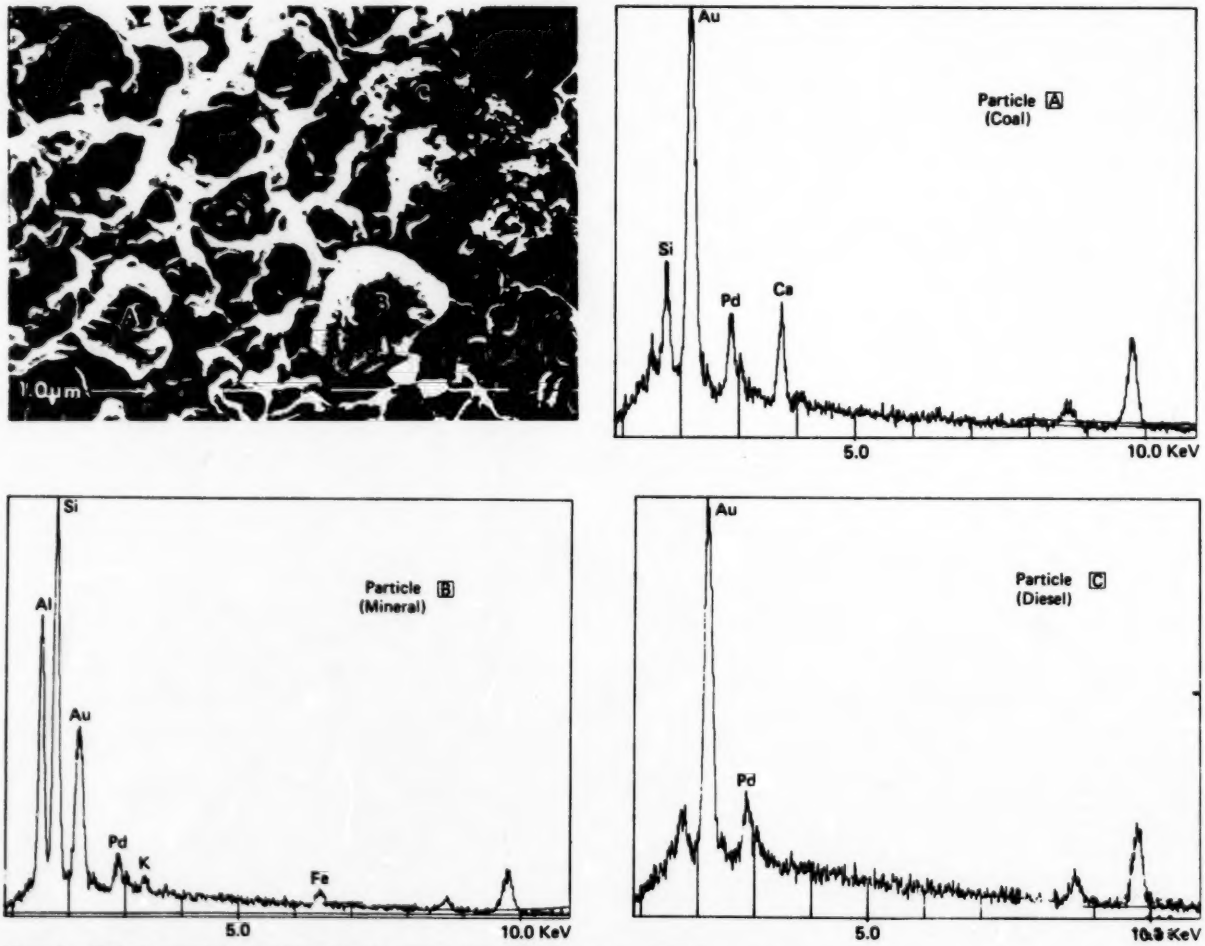


Figure 5. SEM micrograph showing particle types sampled at the strip mine and associated X-ray spectra.

level. Flight operations were conducted at downwind distances 300 to 1000 m from selected mine operations with sampling heights up to 120 m.

Millipore and Nuclepore filters were used as collection substrates during this study. Sampling times were typically 4 to 6 hr which proved to be adequate for optical and scanning electron microscopy. The samples were analyzed for size distribution, number concentration, and composition vs. sampling height.[27]

The wind vane samplers were also used to sample fly ash from the plume of a power plant where the sampling distance was too close (<100 m) for the balloon to be employed directly in the plume. During this field operation the balloon was flown above the plume while wind vane samplers, attached to the tetherline, were positioned in the plume.

To date, all balloon sampling has been conducted during the daytime and most field operations have taken place when the wind was predominantly from one direction. When winds shifted while sampling the plume from a point source emitter, the balloon was moved into the plume simply by moving the winch to a new location directly under the plume, provided no obstructions were in the balloon's flight path. Care was always exercised to ensure that the balloon was operated at safe distances from transmission lines and buildings.

When field operations were conducted over several days, the balloon was usually kept inflated for the entire period. If possible, the inflated balloon was stored in an enclosure during nonsampling periods to ensure that it was not damaged by high velocity winds or strong turbulence.

A 16 mm movie camera equipped with an intervalometer was used to take time lapse movies of all tethered balloon sampling operations. Time lapse photography has proved to be extremely useful in documenting the vertical placement of the balloon in visible plumes, activities at site being sampled, and general atmospheric conditions during flight operations.

Sample Analyses

Although lightweight tethered balloon sampling systems are restricted to the collection of relatively small samples, the types of analyses that can be applied are numerous. Analyses conducted in our laboratory on particles collected from the plumes of point and nonpoint sources have consisted of: (1) scanning electron microscopy/energy dispersive X-ray spectrometry (SEM/EDXS), to determine particle size distributions, concentrations, morphologies, and elemental compositions; (2) transmission electron microscopy/selected area electron diffraction (TEM/SAED), to determine the species of individual particles; and (3) X-ray fluorescence (XRF), to determine major, minor, and trace elements of bulk samples. The bulk samples were also suitable for atomic absorption, neutron activation, proton induced X-ray emission, and X-ray diffraction analyses.

Optical microscopy has also been employed on dust particles greater than 2 μm to determine size distributions and concentrations of particles with different optical properties.

The tethered balloon sampling systems are also well suited for collecting samples for *in situ* microchemical analyses where the collected particles: (1) react directly with the collection substrates; (2) react directly with a chemically coated substrate, or (3) react later upon humidification of a chemically coated substrate in the laboratory.

Methods have been developed to detect atmospheric sulfuric acid droplets collected on thin metal films. By either analyzing the spots formed by chemical reactions on the film surfaces or the reduction in film thickness, it is possible to measure accurately the concentrations and size distributions of the droplets.[32,33] Another microchemical analytical method involves the collection of particles on a transmission electron microscope substrate covered with a thin layer of vacuum evaporated barium chloride. When sulfate particles are present and the substrate is humidified, a reaction occurs that identifies the presence of individual particles containing sulfate ions and the amount of sulfate.[34,35] A less commonly used technique involves the use of Nitron, a chemical used to identify nitrates in bulk analysis. When this material is vacuum deposited on a TEM substrate, it provides a means of identifying individual nitrate and sulfate particles which produce different but distinctive patterns when humidified.[36]

Examples of Results

An example of an SEM/EDXS analysis is illustrated in Figure 5. Shown in the figure is a SEM micrograph of particles collected during the surface coal mine study.[27] The relative elemental composition of each particle, as analyzed by EDXS, is also shown. The gold peak recorded in each sample spectrum is produced by a 10 nm thick gold/palladium alloy coating. Prior to the analysis, the coating is vacuum sputtered onto the filter samples to make them electrically conductive for high resolution electron beam observations. It is notable that coal particles often contain significant amounts of elements normally associated with minerals (Al, Si, Ca) whereas the carbon black-type particle, probably diesel, produced no observable X-ray spectra. Carbon black particle agglomerates were only rarely observed in the coal mine samples examined. In general, particles having a gold peak the same as or less in magnitude than Si (or other major elemental peaks) were classified as mineral in this analysis.

Particle counting to determine size distributions and concentrations of sampled coal and mineral particles was performed from SEM micrographs. A number of micrographs of randomly selected fields at magnifications of 80X and 640X were examined for each sample. The composition of the particles was noted directly on the micrographs prior to counting.

All particles observed on the micrographs were classified by size and composition and tallied into size classes based upon a geometric progression. After classifying the mineral and coal particles separately, the particles counted per interval at 640X were extrapolated to the number which would be present at a magnification of 80X. For intervals which overlapped the two magnifications, weighted averages of the number of observed particles were employed. The total number of particles in each interval across the entire active filter was then calculated and, in turn, divided by the total volume of air sampled through the filter. The mid-size diameter of each size interval was used to calculate a spherical particle volume. These volumes were then multiplied by the appropriate number counts per volume of air sampled and by assumed particle densities so that cumulative and differential mass distributions as functions of particle size could be plotted. The assumed density of the mineral particles was 2.6 g/cm^3; that for the coal particles, 1.4 g/cm^3.

Distributions for particles sampled at a height of 10 m and a downwind distance of 400 m from a number of mining activities are shown in Figure 6. The straight lines through the data points of the cumulative mass distributions were fitted using a least squares routine applied in log-normal space. If a distribution is log-normal, it will occur as a straight line when plotted on a log-probability graph. In general, the data are approximately log-normal. The mass median diameters, d_{gm}, are the 50% values of the distributions while the geometric standard deviations, σ_g, are the 84.1% values divided by the 50% values. For this sample, d_{gm} values for mineral and coal particles are 18.1 and 21.1 μm, respectively, while σ_g values are 2.4 and 2.6.

The differential mass distributions are log-log plots of the mass per interval divided by the difference of the log D interval boundaries (μg/m^3) vs. particle diameter (μm). The data points have been joined by smooth curves using a

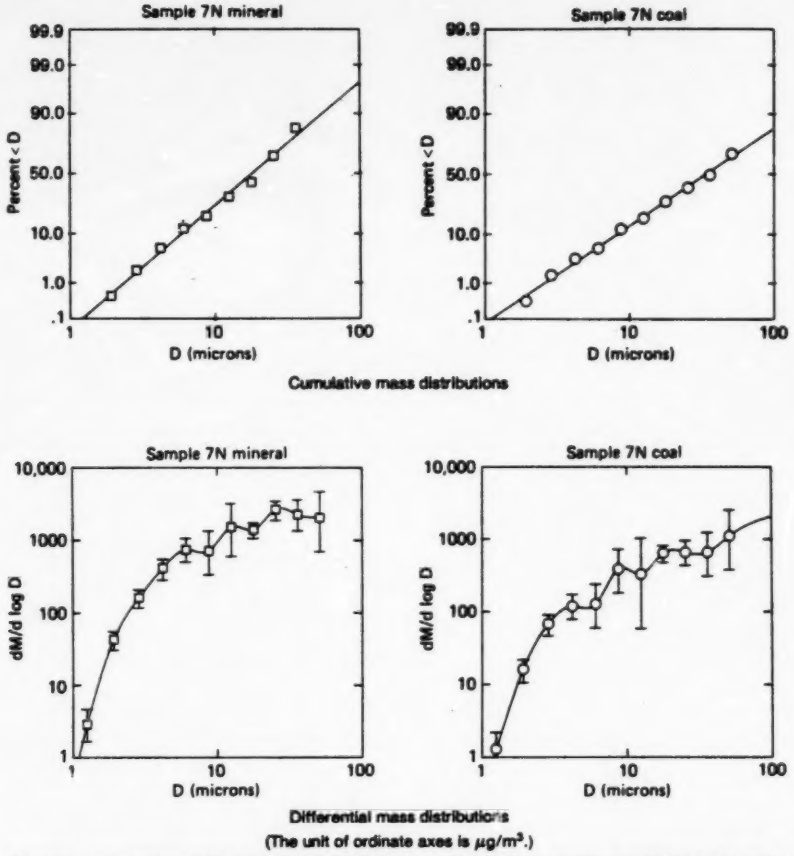


Figure 6. Cumulative and differential mass distributions of mineral and coal particles sampled at an elevation of 10 m.

spline fit routine. The error bars are 90% confidence limits for a Poisson distribution. The larger error bars at a particle diameter of ~12 μm for both the mineral and coal distributions are due to the fact that the 80X and 640X magnifications used in sizing were sufficiently far apart at the size overlap that fewer particles were counted in this size range.

Another example of an SEM/EDXS analysis involves the examination of materials collected from a high-opacity persistent plume of a Portland cement plant.[26] The persistent plume was always associated with the prior formation of a water condensate plume which at times would be detached from the stacks of the plant's electrostatic precipitator. When the ambient air temperature was high enough that the water condensate plume did not form, the plume was clear. Prior to the balloon sampling, the composition of the persistent plume was unknown although a number of in-stack sampling programs had been previously conducted.[37,38] The persistent plume samples revealed that the dominant particles by number had submicrometer diameters and were observed to be of a liquid nature, when examined by SEM with the sample substrate at a high tilt angle to the normal electron beam axes.

An examination of the individual liquid particles by EDXS showed that they were rich in sulfur. These particles were observed to be stable in all but the most intense SEM beam, where some decomposition was observed. The liquid morphology of these sulfur-rich particles strongly suggested that surfuric acid was the dominant component. H_2SO_4 possesses an extremely low vapor pressure and will exist as a liquid in the vacuum necessary for SEM analysis.[39,40]

TEM and SAED analyses were performed which confirmed that the sulfur-rich particles were not crystalline. Further laboratory testing involved exposing the samples to ammonia vapor which caused stable crystals to form.

In-stack sampling was conducted concurrently with the plume sampling. The stack gases contained about 20% water vapor and average SO_2 and NH_3 concentrations of 200 and 170 ppm, respectively.[41] Based upon the laboratory observations of the plume samples and the in-stack gas measurements, it has been proposed that the liquid particles from the cement plant consisted of H_2SO_4 most likely containing some dissolved $(NH_4)_2SO_4$ and/or NH_4HSO_4, created by the aqueous oxidation of SO_2 in the presence of NH_3.

Microchemical analyses of individual particles have been conducted on fly ash collected from the plume of a coal-fired power plant. The particles were collected directly onto TEM substrates using a lightweight balloon-borne impactor.[25] The analyses employed are based upon the observations made by TEM of the specific reaction of sulfate ions with a thin layer of a suitable reactant deposited on the collection substrate and adhered particles. Two reactive materials, $BaCl_2$ and Nitron, have been used following the techniques described by Mamane.[35,36]

Conclusions

A number of lightweight tethered balloon samplers have been developed and utilized by DRI. Their use demonstrates the feasibility and versatility of the tethered balloon sampling approach which allows rapid horizontal and vertical mobility as well as near-source and longterm sampling. Emissions from both point and nonpoint sources have been sampled and analyzed where ground, tower, or aircraft sampling would have been inadequate or impractical.

Areas of environmental interest where the tethered balloon sampling technique could prove to be particularly valuable include:

1. The study of condensation and/or chemical reaction gas-to-particle conversion phenomena that occur exterior to the emission source. These particles may be either liquid or solid and could be collected on chemically neutral or specifically reactive substrates suitable for bulk and microanalytical examinations.
2. The establishment of improved emission factors from fugitive emission sources utilizing a network of balloon systems in conjunction with the exposure profiling technique.
3. The collection of long-term samples from multiple sources, such as urban air masses, especially during periods of atmospheric inversions.
4. The collection of adequate plume samples for bioassays of airborne material.
5. The bulk and microcharacterization of pollution sources for source/receptor chemical mass balance modeling. The sampling technique could also provide important information on intermediate product

formation between the source and receptor.

Acknowledgment

This work was sponsored by the U.S. Environmental Protection Agency under Grant No. R804829.

References

1. A. H. Howell, "Background on Balloon-Borne Experiments," in *Proc. Eighth AFCRL Scientific Balloon Symposium*, AFCRL-TR-74-0393 (NTIS AD-A003398), August 1974. pp. 425–432.
2. C. J. Readings, H. E. Butler, "The measurement of atmospheric turbulence from a captive balloon," *Meteor. Mag.* **101**: 286 (1972).
3. D. A. Haugen, J. C. Kaimal, C. J. Readings, A. J. Marks, "The Minnesota 1973 Atmospheric Boundary Layer Experiment," in *Proc. Eighth AFCRL Scientific Balloon Symposium*, AFCRL-TR-74-0393 (NTIS AD-A003398), August 1974, pp. 89–97.
4. A. L. Morris, D. B. Call, R. B. McBeth, "A small tethered balloon sounding system," *Bull. Amer. Meteor. Soc.* **59**: 964 (1975).
5. C. D. Whiteman, T. B. McKee, "Observations of vertical atmospheric structure in a deep mountain valley," *Arch. Met. Geoph. Biokl., Ser. A* **26**: 39 (1977).
6. C. D. Whiteman, T. B. McKee, "Air pollution implications of inversion descent in mountain valleys," *Atmos. Environ.* **12**: 2151 (1978).
7. G. R. Hilst, N. E. Bowne, "Diffusion of aerosols released upstream of an urban complex," *Environ. Sci. Technol.* **5**: 327 (1971).
8. P. Mandrioli, F. Tampieri, "Vertical profiles of biological particle concentrations under convective conditions," *Boundary-Layer Meteor.* **14**: 331 (1978).
9. I. Galbally, "Some measurements of ozone variation and destruction in the atmospheric surface layer," *Nature* **218**: 456 (1968).
10. K. Sekihara, Y. Suzuki, I. Akita, T. Suzuki, W. Yagihashi, "Ozone concentration in the lower atmosphere," *J. Meteor. Res., Tokyo* **29** (5–6): 105 (1977).
11. G. A. Cleeves, T. J. Lemmons, C. A. Clemons, "A low-level air sampling and meteorological sounding system," *JAPCA* **16**: 207 (1966).
12. L. Rohrschneider, A. Jaeschke, W. Kubik, "Examination of air impurities at heights up to 500 m above industrial sources," *Chem. Ing. Techn.* **43** (18): 1010 (1971).
13. P. Perroud, M. Sylvestre-Baron, G. Pleyber, P. Perilhon, R. Faivre-Pierret, A. Closson, C. Nicotra, "Air Pollution Measurement in the Lower Layers of the Atmosphere. The Use of Captive Balloons," presented at the Colloquium on Pollution Control Techniques, Grenoble, France, October 2–6, 1973.
14. P. Perroud, G. Pleyber, M. Sylvestre-Baron, "Use of Captive Balloons for the Study of the Pollution in the Lower Layers of the Atmosphere," (NTIS CEA-CONF-2491), presented at the Aerall Colloquium, Paris, France, November 12–14, 1973.
15. J. A. Garland, J. R. Branson, "The mixing height and mass balance of SO_2 in the atmosphere above Great Britain," *Atmos. Environ.* **10**: 353 (1976).
16. P. W. Ryan, C. D. Tangren, C. K. McMahon, "A Balloon System for Profiling Smoke Plumes from Forest Fires," Paper No. 79-6.1, presented at the 72nd Annual Meeting of the Air Pollution Control Association, Cincinnati, OH, June 1979.
17. R. J. Sentell, R. W. Storey, J. J. C. Chang, S. J. Jacobson, "Tethered Balloon-Based Measurements of Meteorological Variables and Aerosols," NASA TMX-73909 (NTIS N77-15586), December 1976.
18. J. M. Rosen, D. J. Hofmann, "Investigation of Near Surface Aerosol Properties in Arid Regions," University of Wyoming Final Report No. AP-45, ARO-15199.1-GS (NTIS AD A068554), August 1978.
19. T. F. Heinsheimer, "ATMOSAT, a new measurement platform for air quality monitoring," *JAPCA* **27**: 530 (1977).
20. J. Forrest, S. E. Schwartz, L. Newman, "Conversion of sulfur dioxide to sulfate during the Da Vinci flights," *Atmos. Environ.* **13**: 157 (1979).
21. H. W. Baynton, D. B. Call, N. Medrud, A. L. Morris, J. H. Smalley, "Blimps and Tethered Balloons as Aids to Atmospheric Research," Ad Hoc Committee Report, NCAR Research Aviation Facility Advisory Panel, Boulder, CO, October 1975.
22. M. B. Kuhner, J. R. McDowell, "User Definition and Mission Requirements for Unmanned Airborne Platforms," Battelle Columbus Laboratories Final Report, NASA Contract No. NAS6-2938, December 1979.
23. Federal Aviation Regulations, U.S. Department of Transportation, Federal Aviation Administration, Washington, D.C., Part 101—Moored Balloons, Kites, Unmanned Rockets and Unmanned Free Balloons, August 20, 1974.
24. J. A. Armstrong, P. A. Russell, "The Monitoring of Particulates Using a Balloon-Borne Sampler," in *Symposium on the Transfer and Utilization of Particulate Control Technology*, Volume 4, EPA-600/7-79-044d (NTIS PB 295229), February 1979, pp. 357–376.
25. J. A. Armstrong, P. A. Russell, L. E. Sparks, "The Effects of Conditioning on Near Stack Emissions from a Coal-Fired Power Plant," in preparation.
26. J. A. Armstrong, P. A. Russell, M. N. Plooster, L. E. Sparks, "Tethered Balloon Plume Sampling of a Portland Cement Plant," presented at the Third Symposium on the Transfer and Ultilization of Particulate Control Technology, Orlando, FL, March 1981.
27. J. A. Armstrong, P. A. Russell, D. C. Drehmel, "Particulate Production from Surface Mining - Part 1: Vertical Measurements," in *Proceedings: Fourth Symposium on Fugitive Emissions: Measurement and Control*, EPA-600/9-80-041 (NTIS PB 81-174393), December 1980, pp. 37–63.
28. F. J. Miller, D. E. Gardner, J. A. Graham, R. E. Lee, W. E. Williams, J. E. Bachmann, "Size considerations for establishing a standard for inhalable particles," *JAPCA* **29**: 610 (1979).
29. J. A. Armstrong, D. C. Drehmel, "Balloon Sampling to Characterize Particle Emissions from Fugitive Sources," presented at the Third Symposium on the Transfer and Utilization of Particulate Control Technology, Orlando, FL, March 1981.
30. C. Cowherd, Jr., "Measurement of Fugitive Particulate," in *Second Symposium on Fugitive Emissions: Measurement and Control*, EPA-600/7-77-148 (NTIS PB 276973), December 1977, pp. 47–62.
31. T. A. Cahill, L. L. Ashbaugh, J. B. Barone, R. A. Eldred, P. J. Feeney, R. G. Flocchini, G. Goodart, D. J. Shadoan, G. W. Wolfe, "Analysis of respirable fractions in atmospheric particulates via sequential filtration," *JAPCA* **27**: 675 (1977).
32. S. W. Horstman, J. Wagman, "Measurement of size and concentration of particulate sulfuric acid," *Am. Ind. Hyg. Assoc. J.* **28**: 523 (1967).
33. A. M. Hazrati, "The use of metal films in studying sulfuric acid droplets in the atmosphere," *JAPCA* **29**: 372 (1979).
34. E. K. Bigg, A. Ono, J. A. Williams, "Chemical test for individual submicron aerosol particles," *Atmos. Environ.* **8**: 1 (1974).
35. Y. Mamane, "A Quantitative Method for the Detection of Individual Submicron Sulfate Particles," Ph.D. Thesis, The Pennsylvania State University, Diss. Abstr. 77–23, 253, University Microfilms International, Ann Arbor, MI, 1977.
36. Y. Mamane, R. F. Pueschel, "A method for the detection of individual nitrate particles," *Atmos. Environ.* **14**: 629 (1980).
37. M. Kawahata, "Study on Rotary Kiln Exhaust Gas Emissions in Relation to Blue Haze Formation," Environment/One Corp. Report to Glens Falls Portland Cement Company, November 1974.
38. V. Aggarwal, B. Pine, D. Daoust, H. Potter, B. Gould, T. Clark, "New York State Stack Test Report, Glens Falls Cement Company, Glens Falls, N.Y.," New York State Department of Environmental Conservation Report, September 1975.
39. J. P. Lodge, Jr., "Identification of Aerosols," in *Advances in Geophysics, Vol. IX*, H. E. Landsberg, J. Van Migham (eds.), Academic Press, New York, 1962. pp. 97–139.
40. J. L. Gras, G. P. Ayers, "On sizing impacted sulfuric acid aerosol particles," *J. Appl. Meteor.* **18**: 634 (1979).
41. B. Dellinger, G. Grotecloss, C. R. Fortune, J. L. Cheney, J. B. Homolya, "Sulfur dioxide oxidation and plume formation at cement kilns," *Environ. Sci. Technol.* **14**: 1244 (1980).

Dr. Armstrong is a Research Environmental Engineer and Mr. Russell is a Research Biologist of the Denver Research Institute, University of Denver, Denver, CO 80208. Drs. Sparks and Drehmel are Project Officers, Industrial Environmental Research Laboratory, U.S. Environmental Protection Agency, Research Triangle Park, NC 27711.